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## **JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 52 – 113940 A**

**Title: Solid/Liquid Separation Method**

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(54) **Title of the Invention: Solid/Liquid Separation Method**

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**SPECIFICATION**

**1. Title of the Invention**

Solid/Liquid Separation Method

**2. Claims**

A solid/liquid separation method for an acetic acid slurry of terephthalic acid, characterized in that when paraxylene is oxidized in an acetic acid solvent, the resulting acetic

acid slurry of terephthalic acid is subjected to solid/liquid separation, and the resulting terephthalic acid cake is subsequently washed with acetic acid, the temperature of the acetic acid used for washing is set at 80 to 140°C.

### **3. Detailed Description of the Invention**

The present invention relates to a solid/liquid separation method for acetic acid slurries of terephthalic acid. In further detail, the present invention relates to a solid/liquid separation method for terephthalic acid obtained as a result of the oxidation of paraxylene in an acetic acid solvent.

Polyester fibers have conventionally been produced from dimethyl terephthalate and ethylene glycol, but terephthalic acid and ethylene glycol have been used in so-called direct polymerization over the past few years because of the lower cost of the final product. High-purity terephthalic acid capable of being subjected to direct polymerization is obtained as a result of subjecting the resulting crude terephthalic acid to purification or another type of after-treatment. However, the distillation or other aftertreatment is costly because of the complexity of the process, and the like; therefore, the cost of the terephthalic acid itself increases while the advantages of using the direct method are eventually halved. Consequently, there has been a demand for methods to be studied in regard to manufacturing high-purity terephthalic acid with which terephthalic acid obtained merely as a result of paraxylene oxidation and solid-liquid separation of the product, without distillation or another aftertreatment having to be performed, will be sufficient for use in direct polymerization.<sup>1</sup>

The metal content of the terephthalic acid is generally an important quality of the terephthalic acid used for direct polymerization used to determine the quality of the polyester fibers, and it is necessary this metal content to be reduced to as great an extent as possible. However, when high-purity terephthalic acid suitable for direct polymerization is produced without distillation or another aftertreatment being performed, the metal content that enters the reaction system via the heavy metal catalyst used for paraxylene oxidation and corrosion of the equipment will be present in the terephthalic acid obtained by means of solid-liquid separation. It

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<sup>1</sup> Translator's note: Refer to item 6 of the Procedural Correction on page 7.

has been necessary in the past to subject the terephthalic acid slurry to solid-liquid separation once the reaction has concluded, and then repeat at least twice a procedure to re-slurry the product and re-subject the slurry to solid-liquid separation. Repeating the solid-liquid separation and re-slurrying steps so many times requires that complicated and expensive equipment be used and complicated and highly inefficient procedures performed. When terephthalic acid has been produced with the help of conventional methods, the acetic acid slurry is subjected to solid-liquid separation, whereupon the impurities in the terephthalic acid are washed off with acetic acid. The acetic acid wash is generally performed at standard temperature to 60°C because when a higher temperature is used, the acetic acid will evaporate and cover the interior of the pump used to feed the acetic acid for washing. The pressure must be increased in order to prevent such evaporation from occurring, but increasing the temperature of the acetic acid causes no improvement in the washing effect. The amount of heavy metal catalyst that is used for the manufacture of terephthalic acid from paraxylene in an acetic acid solvent and in the presence of a heavy metal catalyst is very large when compared to other conventional catalyst reactions because the reaction is a liquid-phase reaction, and because the metal catalyst must be present in the form of ions. The terephthalic acid contains approximately 0.1 to 1 wt% of metal in terms of the concentration in the acetic acid slurry. When this terephthalic acid is subjected to one cycle of solid-liquid separation using a centrifuge, and subsequently washed with acetic acid at a common temperature, 20 to 100 ppm of metal will remain in the terephthalic acid. Even if the washing time is prolonged and the other operating conditions are optimized, the heavy metal content will still only be reduced to 20 to 100 ppm in optimal circumstances, and it will be impossible to obtain a metal content of 10 ppm or less, and preferably 5 ppm or less, which relates to the quality demanded for terephthalic acid used for direct polymerization. To a certain extent, it is possible to reduce the metal component and other impurities contained in the terephthalic acid as a result of increasing the amount of acetic acid used for the washing performed after solid/liquid separation has concluded, but if any acetic acid remains after having been used to actually produce the terephthalic acid, the excess will not be able to be used as washing liquid, and a large facility will be required to treat the used acetic acid.

The inventors performed diligent investigations into methods for removing the metal content in terephthalic acid, based on the general industrial condition that the amount of acetic

acid is one to two times the weight of the terephthalic acid that is to be washed therewith. As a result, they ascertained that the distribution of the metal in terephthalic acid obtained by means of liquid-phase air oxidation of paraxylene is lowest in the cores of the terephthalic acid particles, but extremely large at the surface, with 80 to 90% of the metal being deposited near the surface; and that as long as the surfaces of the terephthalic acid particles have not been dissolved as a result of washing, it will be impossible to remove the metal that has adhered to the terephthalic acid. They also discovered that the amount of terephthalic acid dissolved by acetic acid is 85 ppm at 20°C, 150 ppm at 40°C, and 280 ppm at 60°C, but the amount of terephthalic acid that is dissolved by the acetic acid rises dramatically when temperature is 80°C or higher; i.e., 500 ppm at 80°C, 900 ppm at 100°C, and 1300 ppm at 110°C. They also discovered that this dissolution rate is extremely fast, and reaches saturation within several seconds. The present invention was obtained based on the results of this research.

In other words it was discovered that when paraxylene is oxidized in an acetic acid solvent, the resulting acetic acid slurry of terephthalic acid is subjected to solid/liquid separation, and the resulting terephthalic acid cake is subsequently washed with acetic acid, the metal content of the terephthalic acid can be eliminated as a result of bringing the temperature of the acetic acid used for washing to 80 to 140°C.

The acetic acid slurry of terephthalic acid that can be used in the present invention is generally obtained by means of oxidizing paraxylene or a partial decomposition product thereof using molecular oxygen in the presence of a heavy metal catalyst. The slurry contains 15 to 50 wt% of terephthalic acid. The metal content of this terephthalic acid consists of iron and other metals that have been inevitably admixed into the reaction system due to the cobalt, manganese and other auxiliary catalysts that have been used as a heavy metal catalyst and due to corrosion of the reaction equipment. A continuous centrifuge or an automatic batch centrifuge is generally used for subjecting acetic acid slurries of terephthalic acid to solid-liquid separation. The former is used to subject the acetic acid slurry to continuous solid-liquid separation under centrifugal force as the slurry is continuously drawn in to continuously obtain a filtrate and cake, while the latter involves automatic repetition of a cycle whereby a specific amount of acetic acid slurry is drawn in, and the filtrate is shaken off as a result of centrifugation, fresh acetic acid is generally sprinkled to perform washing, the system is shaken, after which the cake is scraped off. These

methods are both used for different purposes, but an automatic batch-type centrifuge is particularly preferred for use the present invention. The automatic batch-type centrifuge generally operates on a cycle in which the slurry is fed, shaken, washed, shaken again, and scraped off. If the filter becomes clogged, it is flushed with an alkali cleaner. The time required for this cycle is within two to five minutes. The terephthalic acid centrifugate is separated from the acetic acid solvent and the acetic acid used for washing. However, centrifugation is sufficient when the acetic acid content reaches 10% or less in terms of the weight of terephthalic acid. The washing liquid temperature is 80 to 140°C, preferably 100 to 120°C, ideally 105 to 115°C. The effect of washing increases as the washing liquid temperature temperature, but the pressure needs to be increased when the washing liquid reaches its boiling point or a higher temperature; excessive pressure will increase the cost of the equipment. Therefore, a pressure of up to 4 kg/cm<sup>2</sup> and a temperature of up to 140°C must be used. As previously described, the amount of washing liquid to be used is preferably one to two times the amount of terephthalic acid in consideration of the equipment and the like, but when the quality of the terephthalic acid needs to be increased further, or when the temperature of the washing liquid is low, the amount of washing liquid to be used can be as much as five times the amount of terephthalic acid. On the other hand, when the terephthalic acid is able to contain a metal component to a certain extent, or when the temperature of the washing liquid is very high, the amount of washing liquid used can be 0.5 time the amount of terephthalic acid. The acetic acid used for washing is not necessarily pure acetic acid, and can contain a certain amount of water or other impurities. When the metal component needs to be completely removed, the washing liquid can be divided into several batches, and the centrifugation and washing steps repeated. The washing liquid will contain a high concentration of terephthalic acid after centrifugation, which can be used untreated as the solvent for paraxylene oxidation.

As previously described, the method of the present invention enables the metal content to be removed from the terephthalic acid, and the metal content in the resulting terephthalic acid to be kept within a range that is suitable for direct polymerization, without complex steps being used, and virtually without any increase in cost in the resulting terephthalic acid.

The present invention will now be described using working examples and comparative examples. References to "%" in the examples designate percent by weight unless otherwise stated.

#### **Working Example 1**

First, 280 kg of an acetic acid slurry (metal concentration: 0.5%) containing 30% of terephthalic acid was fed to a centrifuge over a period of 40 seconds. The mother liquor was shaken off as a result of centrifugation for 40 seconds, and the product was washed for 20 seconds with 120 kg of acetic acid (1.5 times the amount of terephthalic acid) that had a water content of 3% and a temperature of 105°C. The resulting liquid was shaken again for 50 seconds. The cake was scraped off for five seconds and dried with a drier. The metal content of the resulting terephthalic acid was 2 ppm.

#### **Comparative Example 1**

The procedure described in Working Example 1 was performed at a washing temperature of 75°C, with the metal content of the resulting terephthalic acid being 30 ppm. The washing liquid temperature clearly has a strong effect on removing the metal content.

#### **Comparative Example 2**

The procedure was performed in the same manner as described in Comparative Example 1 with the exception that washing was performed for 40 seconds using 240 kg of acetic acid (three times the amount of terephthalic acid), or twice the amount used in Comparative Example 1, with the metal content of the resulting terephthalic acid being 18 ppm. A dramatic effect was obtained as a result of increasing the amount of acetic acid used for washing, but the product was still unsuitable for direct polymerization.

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Procedural Correction (Voluntary)

Date: June 16, 1976

Commissioner, Patent Office Ishiro KATAYAMA

1. Case No. 51-51497

2. Title of the Invention

**Solid/Liquid Separation Method**

3. Party Making the Amendment

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5. Sections Amended

Detailed Description of the Invention of the Specification

6. Amendment Details

"Consequently, there has been a demand for methods to be studied in regard to manufacturing high-purity terephthalic acid with which terephthalic acid obtained merely as a result of paraxylene oxidation and solid-liquid separation of the product, without distillation or



another aftertreatment having to be performed, will be sufficient for use in direct polymerization."

is corrected to:

"Consequently, there has been a demand for methods to be studied in regard to manufacturing high-purity terephthalic acid that will be sufficient for use in direct polymerization merely as a result of paraxylene oxidation and solid-liquid separation of the product, without distillation or another aftertreatment having to be performed" (last sentence of the second paragraph of the section titled "Detailed Description of the Invention").